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Abstract

Chlorination-Distillation Processing of Irradiated Uranium Dioxide and Uranium Dicarbide

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Chlorination-distillation processing of irradiated pellets of uranium dioxide and of uranium dicarbide was studied, with the aim of utilizing the difference in vapor pressure between chlorides of uranium and those of fission products. The condensed uranium chlorides were recovered in the form of dioxide powder by oxidation-reduction cycling.

Pellets were irradiated during either 1 or 4 weeks in the JRR-1 with an intergrated flux of 0.25×10^{16} or 1×10^{16} n/cm^2 and were subjected to experiment after letting the activity decay for a period of between one and five weeks. The dioxide pellets were chemically pulverized by alternately oxidizing in air flow at about 400°C and reducing in hydrogen flow at about 800°C, while the dicarbide pellets were used without pulverization. Carbon tetrachloride was used as chlorinating agent, in the form of vapor flowing through a quartz-tube assembly at various chlorination and condensation temperatures.

It was not possible to obtain the separation of most of the fission products from irradiated uranium dioxide by a simple

process of chlorination-fractional distillation followed by oxidation-reduction, and the decontamination factor (γ) was only about 10 with fuel cooled 40 days. The γ-emitting nuclides remaining with the recovered uranium were mainly ^{141}Ce, ^{144}Ce and ^{239}Np. ^{239}Np decays rapidly due to its short half-life. In order to separate the radioactive cerium, natural cerium dioxide powder was added to an amount corresponding to 2 o/o of the irradiated uranium dioxide before chlorination-distillation, which was followed by the oxidation-reduction cycle. The decontamination factor obtained in this case was approximately 100 with fuel cooled 40 days. By repeating this process with cerium dioxide addition, the decontamination factor was further raised to about 400, with more than 97 o/o of uranium recovered.

The uranium dicarbide pellets were directly chlorinated without pulverizing, and distillated fractionally immediately after chlorination. Carbon remained involatile, retaining most of the cerium. The decontamination factor (γ) obtained was approximately 150 on fuel cooled 40 days. For better decontamination, a second chlorination-fractional distillation was conducted after adding 2 o/o cerium dioxide, followed by the oxidation-reduction cycle, as on the irradiated uranium dioxide. The decontamination factor of the recovered uranium dioxide was about 900. More than 95 o/o of uranium was recovered.

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Abstract

Recent Development of Beryllium in Japan

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This paper describes the outline of the recent research and development of beryllium metal and beryllium oxide for nuclear reactor materials with special reference to fuel matrix or fuel cannings and other reactor components for the reactor with high temperature and high burn up.

1. Trends of Research and Development:

Research and development of beryllium-base materials for nuclear reactor use is believed to be important from the point of view of nuclear fuel situation of this country, that is, it is better to avoid to use enriched uranium fuel as possible as it can be. This research and development also must be necessary for development of nuclear reactor with high temperature and high burn up.

From another point of view, the demands for beryllium metal is not so big, and so its price is rather high. Therefore, great interest has been indicated to the utilization of beryllium oxide which is cheeper than the metal, as nuclear reactor material.

Research and development of the utilization of beryllium oxide should be continued more and more as well as the efforts to produce cheeper beryllium metal.

2. Research of Fubrication of Beryllium Pipe by Means of Extrusion Method:

Beryllium metal has been considered as one of the best materials for fuel cannings for the reactor of natural uranium - Co_2 cooled type.

As the first step of the preparation of fuel cannings, fabrication of bellium pipe by extrusion was investigated and the high temperature properties were also examined by high temperature hardness testing.

Electrolytically prepared beryllium (Boron content less than 2 ppm) was melted in vacuum to remove chlorine that was contained in original flaky beryllium with the amount of 2000 ppm approximately. By this treatment, it was reduced to about 50 ppm.

Beryllium billet for extruding prepared by the process of vacuum melting-powdering-vacuum hotpressing-machining was fully sheathed by mild steel. After preheating the billet in fused NaCl - 36 mol % $BaCl_2$ to 1150°C, it was placed into billet container and then extruded with the extrusion ratio of 1/4.

The effect of purity of beryllium on the extrusion characteristics was checked, but remarkable deference was not observed.

Tab. 1 Chemical Analysis of Beryllium Flake Used in the Experiment (ppm)

	Fe	Al	Cu	Ni
Higher grade	350	90	70	140
Lower grade	2300	300	280	300

Tab. 2 Tensile Properties of Extruded Beryllium (Longitudinal direction, as extruded)

Tensile Strength	Elongation
44 kg/mm^2 (60,000 lb/in^2)	12 %

Longitudinal tensile properties as extruded state was shown in tab. 2. Ductility in this direction was sufficient considerably.

As the results of high temperature hardness measurement, no remarkable plateau in temperature-hardness ourve was appeared.

R. Thomson et al - J. Lesscomm Metals, 3 (1961), 170 - reported the appearance of plateau in extruded bar of thermally reduced pebble beryllium, and Kaufmann - Trans. ASM, 42 (1950), 785 - showed the decrease of elongation in the temperature range between 400 to 800°C in the same material.

Supposedly our extruded and anneal beryllium may show no decrease of elongation in this temperature range. This problem is now under investigation as well as transverse ductility problem and these details will be obtained in the near future.

3. Recent Advances in Beryllium Oxide:

Most efforts of research has been directed to develop fuel matrix material and reflector or moderator material in beryllium oxide field.

3.1. Highly Sinterable Beryllium Oxide Powder:

High purity and high sinterable beryllium oxide powder called CF-grade BeO by the process of some combination of recrystallization and thermal decomposition of beryllium sulphate and fabrication of shapes of this powder have been investigated.

Tab. 3 Relationship between Sintering Temperature and Density of CF-grade Beryllium Oxide Powder and Conventional Powder

Powder Grade	Sintering Temperature	Density
CF	1400°C	greater than 2.90 g/cm^3
CF	1500°C	greater than 2.95 g/cm^3
CF	1600°C	greater than 2.98 g/cm^3
Conventional	1600°C	2.80 ~ 2.90

This powder was more sinterable than conventional powder as shown in tab. 3.

Boron and Lithium content of this powder is below 0.5 ppm, and below 0.5 ppm respectively and so that chemical and nuclear purity were both very high. Therefore, this is sufficient for nuclear use. Particle size of this powder was estimated to be finer than conventional powder (0.1 ~ 0.2 μ) by electron microscope and BET method.

3.2. BeO - Be Shapes:

Mechanical strength as well as the superiority of nuclear and thermal properties is important to utilize beryllium oxide shapes as nuclear reactor materials.

This study was intended to develop high strength beryllium oxide shapes.

High purity beryllium oxide powder with 0.1 to 5 wt % of metallic beryllium powder was hot pressed in vacuum at the temperatures of 1200 to 1500°C, and the relationships among sintering condition, composition, and characteristics of sintered body was investigated and BeO-Be shapes with bending strength of 2000 to 3000 kg/cm^2 (28,000 to 43,000 lb/in^2) and density of 99% up of theoretical or more was easily obtained.

This value of bending strength of BeO-Be shapes was higher than BeO mono-component shapes of the same density. Besides, thermal expansion coefficient and thermal conductivity were approximately equal to those of BeO mono-component shapes.

This BeO-Be can be considered as reflector or moderator material and fuel matrix material (BeO-Be-UO_2 & BeO-Be-ThO_2U_2) especially for high temperature reactors.

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Abstract

DOUBLE REACTOR CONTAINMENT MEETS THE SITE REQUIREMENTS FOR A CIRCULATING LIQUID FUEL REACTOR

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The 250 kW Kema Suspension Test Reactor, a circulating liquid fuel reactor, is under construction on the KEMA-grounds near Arnhem, the Netherlands. The principle of double reactor containment is applied to meet site requirements concerning dose limits at zone boundaries as used by the U.S.A.E.C.

The reactor system (first barrier between the suspension and the outer atmosphere) is placed in steel compartments, backed up by the concrete of the biological shield (second barrier, first reactor containment), which in turn are placed in a concrete reactor hall (third barrier, second reactor containment).

The contingent release of radioactive gases to the environment will be considered for 4 cases, viz. normal operation, maintenance, a major accident, and a hypothetical accident.

The maximum concentration of radioactivity in the gases discharged from the 25 m high stack is set by the authorities to be 10^{-7} curie/m^3 for design purposes. To maintain the underpressure in the nitrogen filled compartments during normal operation, gases are discharged through the stack via charcoal adsorption beds for delay and will always pass absolute filters and iodine traps.

For maintenance purposes after shut down of the reactor the compartments have to be opened. Beforehand they have to be practically free of radioactive gases that might have leaked from the reactor system. This is achieved by a combination of decay and purging. To complete this within reason-

able time the concentration of radioactivity in the stack effluent has to exceed 10^{-7} c/m^3 for short times. This concentration will not reach the value of 10^{-4} c/m^3, being the limit at which the valves in the stack and the ventilation inlet duct will close automatically.

In the case of a major accident (release of the reactor content into the intact compartments) this limit is reached and the valves are closed. The arguments that the integrity of the first containment remains intact are discussed. Decay and controlled release via an emergency charcoal adsorption bed make it possible to remove practically all of the radioactive gases from the compartments in about 3 months without violating the site requirements.

Even for a hypothetical accident in which all fission gases (700 kc) are released into the reactor hall, the site requirements can be met.

To show this one of the points to be considered is the meteorological dilution between the stack and the dose receptor. Due to the small distances to inhabited regions, also the concentration of radioactivity above ground level has to be considered. For a vertical plane through the stack and the wind direction a graph, based on Sutton's formula, was constructed to locate the point of maximum concentration in these inhabited regions.

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Abstract

TWO-PHASE FLOW INVESTIGATIONS FOR A MARINE BOILING WATER REACTOR

by

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The paper describes research work done in support of a marine nuclear reactor design project. It is important to be able to predict the dynamic behaviour of a boiling water reactor in a seaway. Since only the hydraulic circuit is affected, it is sufficient to investigate this experimentally, using the experimental results as a partial check on the validity of a mathematical dynamics model for the whole reactor.

Tests have been done in a 30 kW, 50 ata natural circulation loop which can be oscillated vertically. The stroke is 10 meters, and the accelerations are between 0.6 g and 2 g.

The momentum of the circulating water is important for the dynamic response of a boiling water reactor. A design is described which gives the designer freedom to vary the inertia of the water in the downcomer within wide limits. The effect of this design on the dynamic behaviour of the reactor is described in the paper by H. Christensen et al. The results indicate that powerful circulation pumps are unnecessary.

However, forced circulation improves the stability of a boiling water power reactor. Instead of conventional pumps it is proposed to use a combined steam injector-jet pump, which gives negligible expenses compared with conventional pumps. The injector theory will be explained, and experimental data from a loop with maximum pressure 100 ata and maximum power 300 kW will

be reviewed. The influence of the injector on the B.W.R. design and behaviour will be discussed.

In a reactor using natural circulation or injector circulation the circulation is greatly affected by carry-under of steam into the downcomer, which reduces the natural circulation driving head available. It appears that carry-under must be limited to very small values in such reactors.

Carry-under has been investigated experimentally in the mobile loop referred to above, and in large-scale air-water loops. By regarding carry-under as an aspect of a two-phase flow pattern problem, it has become clear that geometrical effects are important. These have been investigated and the results are given.

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Abstract

NUCLEAR POWER IN KARACHI ELECTRIC SUPPLY SYSTEM

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The Power Commission of Pakistan has come to the conclusion that over the next few decades there will be a large gap between demand and supply of electricity that can be generated from indigenous conventional sources so far discovered. Nuclear power therefore is the only alternative source which can be expected to fill this gap gradually on a long-term basis. The only indigenous fuel available at Karachi is natural gas which is supplied through a 16" pipeline, the delivery capacity of which will soon be saturated if power gaps are met by gas-fired stations. It has therefore been decided to establish a 132 MW natural uranium fueled heavy water moderated and cooled reactor, scheduled to be commissioned by 1969, fourteen miles west of Karachi. The total generating capacity of Karachi system at that time will be 335 MW with a peak demand of 300 MW, which includes power requirements of a steel mill likely to be in operation by that time. The economic feasibility studies indicate that under certain financial terms usually obtainable for power projects in Pakistan the cost of generation offered from the nuclear plant would be lower than from a gas-fired station of equipment capacity for plant factors up to 70% and above. As it is intended to operate the nuclear plant as a base load station the plant factor is expected to be higher. Although the construction cost of nuclear power stations is almost double the cost of a gas-fired station of equivalent capacity, savings on fuel, which are expected to be of the order of $2 million per year, will result in net savings on the operation of the plant of the order of $1 million per year.

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Abstract

TRANSIENT AND STEADY STATE SOLUTIONS OF HEAT TRANSFER THROUGH SPIRAL FINS

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A theoretical treatment is given of transient and steady state heat transfer through spiral fins for application in reactor fuel rods. The heat transfer equation is made dimensionless and solved by the method of Schmidt and Collatz. The solution is more rigorous than the approximate approach of Katayama and Kawashimo.

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Abstract

THE ROLE OF DILUENT IN EXTRACTION PROCESSES

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In many practical extraction processes suitable diluents have been used with the aim to improve the physical properties of the organic phase employed.

Although these diluents were selected from the solvents which are chemically inert, nevertheless they are taking a more or less, active part in the extraction process, with influence upon the partition coefficients. In previous studies carried out in this Institute it was suggested that an appropriate approach to this problem must take into account two independent chemical interactions in the organic phase influencing the extraction equilibrium. These are: the interaction between the extracting agent and the diluent and the interaction between the extracted species and the diluent. To understand the overall effect of the diluent, at least one of these interactions should be measured directly, the other can then be calculated from the measured partition coefficient. The most convenient procedure is to determine the thermodynamic activity of the extracting agent as a function of the composition of the organic phase. This can be done by studying the partition of the extracting agent between the organic phase and an aqueous phase of constant composition. The thermodynamic activity of the extracting agent easily can be related to the mode of interaction with the diluent.

The suggested method is of general applicability and was previously successfully used in the study of the TBP and amine extraction systems. Since acidic extracting agents become more and more important and very probably may find application in fuel reprocessing, it seemed to be worth of applying this method to such systems.

In the experimental part of this paper some recent results are presented on the extraction of europium with dibutylphosphate /DBP/. According to the method discussed above, first, the thermodynamic activity of DBP in seventeen diluents was determined by measuring the partition of this extractant between the organic and aqueous phase. From the measured activity of the extracting agent, and from

the europium partition data obtained for the same concentration of DBP, the magnitude of the interaction between the Eu-DBP complex and the diluent was calculated in the form of the thermodynamic activity coefficient. The interaction with the diluent was explained in part by the application of the concept with regard to the solubility parameter as a measure of the intermolecular interaction involved. The important role of interaction through hydrogen bonding of both the extractant and the complex with some diluents was revealed.

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Abstract

CHLORINATION AT ELEVATED TEMPERATURES AS A VERSATILE URANIUM ORE CONCENTRATION METHOD

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With the aim to find more economic uranium ore concentration methods attention has been drawn to the possibility of chlorine attack at elevated temperatures. New, detailed research work has been performed concerning the reaction of chlorine with uranium oxides and uranium minerals at different temperatures and this with pure chlorine as well as in presence of reducing agents: carbon and carbon monoxide. The mechanism and kinetics of the reactions has been studied with the aid of the radioisotope Cl-36 acting as a tracer. This research work revealed an important fact that the reaction of uranium dioxide with chlorine proceeds already at 225°C, uranyl chloride UO_2Cl_2 being formed, while chlorination of U_3O_8 as well as of UO_3 in presence of carbon preceeds at 300°C.

The results thus obtained may be used as a basis to develop different technological schemes which may be adapted to different ore types. The decisive role is played by the content of other valuable elements contained in ores which form volatile or extractable chlorides in the same process /e.g. titanium, vanadium, molybdenum, niobum and tantalum/. Possibilities for anhydrous aluminum chloride and silicon chloride production in the same process also exist, both being valuable by-products.

A new, high-efficient apparatus to perform reactions of granular solids with gases at elevated temperatures has been developed.

Chlorination is considered to be a general, intense and versatile uranium ore processing method.